Bending Behavior of Monolithic Ba(Ti,Sn)O_3-Ceramics with a Functionally Gradient of the Piezoelectric Properties

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Abstract

Functionally Gradient Materials are suitable for bending devices due to reduced internal mechanical stresses and lower production costs. Monolithic $BaTi_{1-x}Sn_xO_3$ (BTS) ceramics with a gradient of the Sn-content ($0.075 \le x \le 0.15$) were prepared. The chemical gradient was transformed into a gradient of the piezoelectric properties by a poling process.
The bending deflection was measured as a function of the applied voltage and compared with analytical approximations. Additionally, the resonance frequencies and the damping behavior of samples with different Sn-contents were investigated. The monolithic ceramics showed good results in comparison with conventional glued bending devices.

Introduction

The bending effect of piezoelectric bimorph actuators is used in applications where large displacements are required. Bending actuators are usually designed as unimorph with one active piezoelectric layer or bimorph with two active layers bonded by glue [1]. The life time and reliability of conventional bending actuators are reduced by cracks and peel off at the interface between the ceramics and the glue due to high mechanical stresses. Piezoelectric Functional Gradient Materials (FGM) are suitable to reduce this internal stresses and improve the life time of an actuator [2]. They can be easy integrated in smart material applications.
Bending actuators with a piezoelectric gradient are successfully prepared with ceramics from the PNN-PZT system [2, 3]. This material are well-known for their huge piezoelectric coefficients. But for large bending deflection it is not necessary that the material has high electromechanical coefficients. Only the difference in the strain in the length direction S_1 between neighboring layers is important. Usually, in Functionally Gradient Materials the gradient of the piezoelectric properties is coupled with a gradient of the dielectric coefficient of the ceramics. Because a one dimensional gradient is prepared along the thickness, perpendicular to the electric field, the layers with different dielectric properties were serial connected. Thus, the induced electric field is than higher than lower the dielectric coefficient in this layer. An ideal bending actuator should have at one side high piezoelectric coefficient d_{31} and low dielectric permittivity ε_{33} (large strain S_1) and at the opposite side low piezoelectric and high dielectric coeffcients (low strain).
In the last years $BaTiO_3$ ceramics become more and more the aim of interest for developing lead free ceramics for smart materials. In the system $BaTiO_3$-$BaSnO_3$ the Curie temperature decreases with decreasing tin content. The material coefficients change in a wide range around the phase transition. At room temperature the piezoelectric coefficients decrease and the dielectric coefficient increases with increasing Sn content from 7.5 up to 15 mol%.

Sample preparation

Monolithic $BaTi_{1-x}Sn_xO_3$ (BTS) ceramics with a gradient of the tin content were prepared by successive pressing a number of layers of BTS powders with different compositions produced by a classical mixed-oxide technique. To influence the tin gradient 2, 3 or 4 layers were used whereas the whole thickness of the sample was always about 1.1-1.2 mm. The outer layers consist of $BaTi_{0.925}Sn_{0.075}O_3$ and $BaTi_{0.85}Sn_{0.15}O_3$, in the following named BTS7.5 and BTS15, respectively. Ba(Ti,Sn)O_3 powder with 10 and 12.5 mol% Sn were used for inner layers.

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The ceramics were sintered in air at 1400 °C for 1 h with a heating and cooling rate of 10 K min^{-1} to obtain coarse-grained ceramics with an average grain size of about 80 µm.

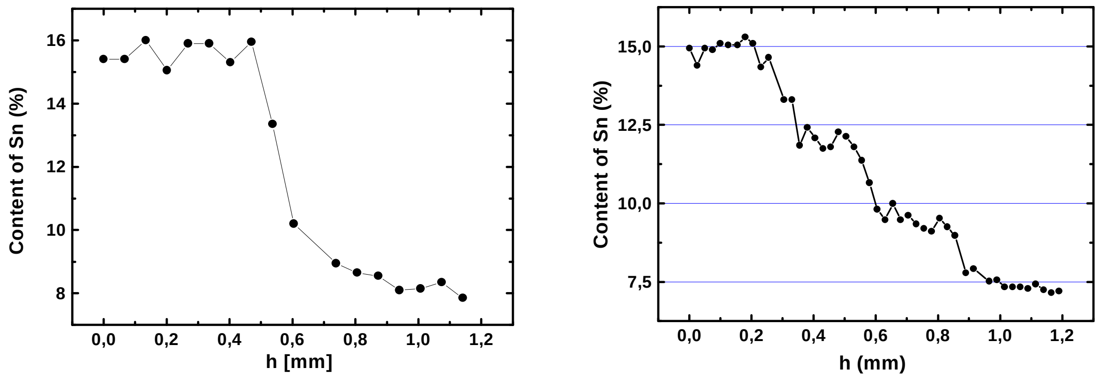

Fig. 1 Content of tin along the thickness of monolithic ceramic samples sintered with two (left) and four layers (right) of BTS

To determine the tin distribution in the ceramic sample along the thickness, wavelength-dispersive x-ray electron probe microanalysis (WDX-EPMA) (model CAMEBAX, Cameca, France) was performed. Figure 1 shows that the chemical composition continuously changes at the interfaces between the layers. On the other hand, the original composition is quite good recognizable. The thickness of the interfaces is about 100-200 µm in dependence on the difference of Sn content between neighboring layers. It should be noted that especially the outer layers with the maximum and minimum amount of tin are relatively thick.
In particular, the monolithic ceramics prepared from four layers (4-morph) have a nearly ideal gradient of the chemical composition. Thus, the terms bimorph, trimorph and fourmorph using in this work are not really correct. These monolithic samples have a gradient of the Sn content. They don‘t consist of layers with a defined thickness, chemical composition and, therefore, defined material properties. The names 2-, 3- and 4-morph only based on the number of powder layers before sintering and as an idealized number of layers for the modeling.
During sintering the monolithic samples bend due to the different shrinking of the $BaTi_{1-x}Sn_xO_3$ ceramic layers. A strong bending effect was obtained for the bimorph structure. Than higher the number of powder layers and than smaller the difference in the chemical composition of neighboring layers than lower the curvature of the sample. The 4-morph structure is nearly unbowed (Figure 2). This effect could be minimized by an additional weight of about 100g laying on the sample during sintering. By this way, unbowed monolithic samples were sintered with only two powder layers (bimorph).
The minimum thickness of each layer prepared by the above described powder pressing is about 0.3 mm. However, the bending deflection is than higher than thinner is the actuator. Thus, thin ceramic foils with a typical thickness of about 80-120 µm were cast. Foils with the appropriate chemical composition were stacked in the required order and sintered under a small pressure. Monolithic ceramics with a whole thickness of 0.3-0.4 mm were prepared by this technology. For bimorph samples two foils with 7.5 and two foils with 15 mol% Sn, respectively, were used.

Fig. 2 Monolithic BTS ceramics sintered with two layers of BTS7.5 and BTS15 (left) and with four layers BTS7.5-BTS10_BTS12.5-BTS15 (right)

After sintering samples were polished and electroded with aluminum electrodes. The gradient of the chemical composition was transformed to a gradient of the electromechanical properties by a poling process. For this, a DC field was applied to the sample for 15 seconds at room temperature. The ferroelectric properties of the BTS ceramics strongly depend on the Sn content. It was shown, that in a ferroelectric multilayer system without

inner electrodes only the layer with the lowest spontaneous polarization will be completely poled. All other layers are only partially poled [4].

To determine the electromechanical and dielectric properties of the poled layers with different Sn content a model structure was used. Single ceramic sheets with the same thickness and chemical composition as the powder layers for the monolithic samples were prepared. These single layers were electroded and electrically connected by a wire. After poling with the same electric field the material coefficients of each layer were measured. The dielectric coefficient ε_{33} and the piezoelectric coefficient d_{31} are used for the modeling of the bending deflection. Subsequently, the single sheets were glued together and the deflection of the glued actuator was determined.

Quasistatic bending behavior

The bending deflection at the free end of a one-side fixed sample was measured by a capacitive displacement sensor [5]. The upper electrode of the specimen is one plate of a measuring capacitance which is a part of a HF-series circuit. The bending deflection of the sample changes the distance between the plates, i.e. causes a change of the capacity of the measuring capacitance and a shift of the resonance frequency of the HF circuit. The frequency shift is transformed into a voltage proportional to the deflection by a modulation analyzer HP8901A.

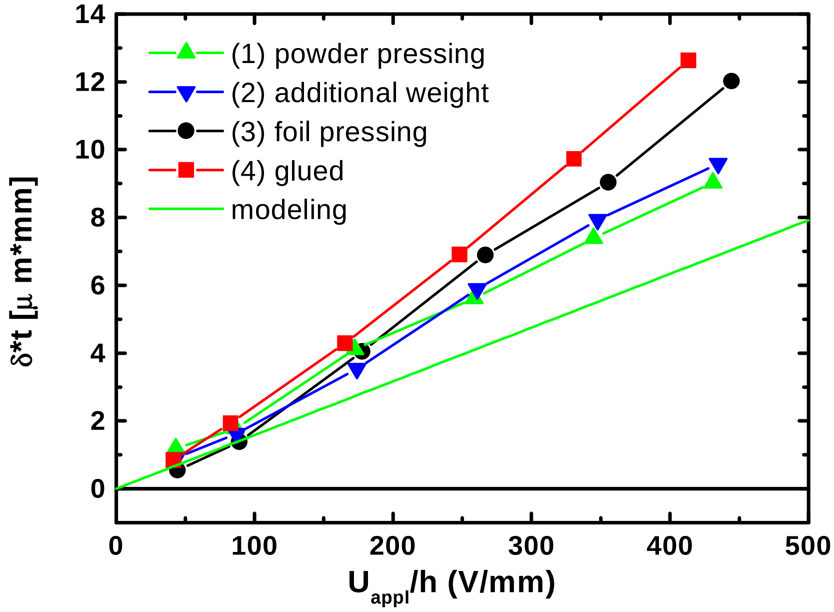

Fig. 3 Deflection of bimorph structures prepared by powder pressing without (1) and with additional weight during sintering (2), foil pressing (3) and conventional gluing of poled layers (4)

An unipolar voltage with a frequency of 137 Hz was applied to measure the quasistatic bending deflection. Figure 3 shows the maximum of the deflection of different bimorph structures in dependence on the applied electric field. The deflection is normalized on the thickness of the sample. The experimental results were compared with an analytical approximation of small signal bending. We assumed, that the elastic properties are independent of the tin content. In this case the deflection δ at the end of the specimen can be calculated by the theory of Marcus [6]

$$\delta = \frac{L^2}{2} \frac{\int_{-h/2}^{h/2} d_{31}(z)\, E_3(z)\, z\, dz}{\int_{-h/2}^{h/2} z^2\, dz}, \qquad (1)$$

where L is the free length and h the thickness of the bending actuator. For small applied voltages U_{appl} the electric field in the layer i depends on U_{appl} and the dielectric coefficients ε_{33} of all layers in the following manner

$$E_3^{(i)}(z) = \frac{U_{appl}}{h^{(i)}}\left(\varepsilon_{33}^{(i)}\sum_{j}^{N}\frac{1}{\varepsilon_{33}^{(j)}}\right)^{-1} . \tag{2}$$

For small electric fields up to 50 V/mm all bimorphs are in a good agreement with the modeling [7]. For higher voltages increases the difference between the experimental results and the linear approximation. On the other hand, Figure 4 shows that the bending loop is relatively linear with a small hysteresis also at higher electric fields.

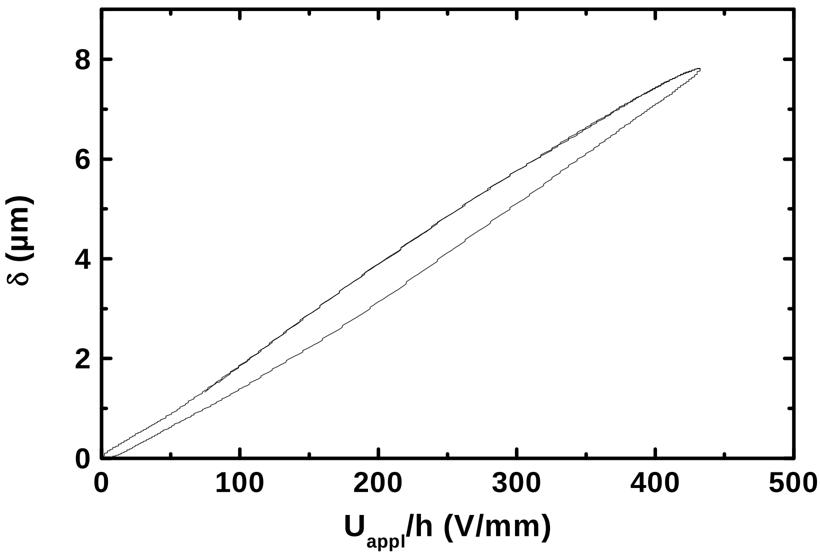

Fig. 4 Bending loop of a monolithic bimorph at 137 Hz

The conventional glued bimorph has the highest value of deflection (Figure 3). The monolithic ceramics with a gradient of the electromechanical properties at the interface of the two materials show a some smaller deflection. From modeling with the Finite Element Method it is known that the deflection of multilayer structures slightly decreases with increasing number of layers [8]. In a first approximation the interface(s) can be considered as additional layer(s).
The additional weight to sinter unbowed ceramics didn‘t significant influence the bending behavior. A slightly higher normalized deflection was obtained at the samples prepared by foil pressing. Much higher absolute deflections at the same electric field can be achieved, if we take into account that the thickness of the specimen is much thinner than the powder pressed ceramics (Eq. 1). Furthermore, the applied voltage is much smaller for the same electric field (Eq. 2). For example, a bimorph with a thickness of 0.4 mm provides a deflection of more than 25 µm at 160 V.

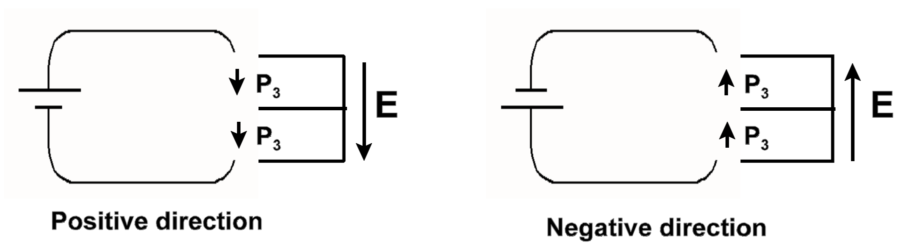

Fig. 5 Definition of the poling direction of the BTS7.5-BTS15 system

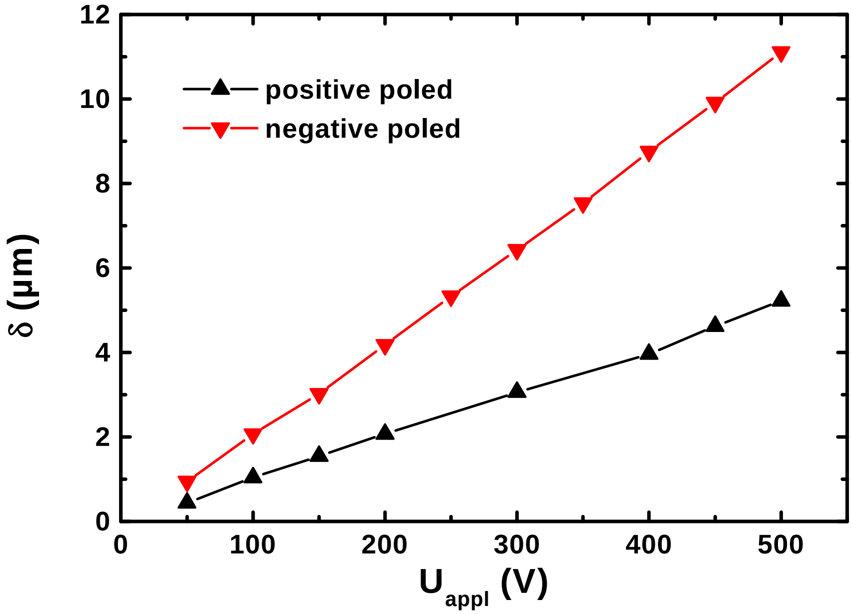

Fig. 6 Deflection of monolithic bimorphs with different poling directions

Influence of the poling direction

Figure 5 shows schematically the two possibilities to apply the poling field to the BTS7.5-BTS15 ceramics. If the positive DC voltage is applied at the BTS7.5 layer we called this sample „positive poled“. „Negative poled“ means that the poling voltage was applied at the BTS15 layer. We obtained a major difference of the bending behavior for the different poling directions. The negative poled monolithic bimorph bends approximately twice more at 500 V than the positive poled one. For comparison we poled the system of electrically connected single ceramic sheets also in the opposite direction. It was found that the BTS7.5 layer (high piezoelectric coefficients) was much better poled in the negative poled system (Figure 6). We assume that the distribution of the electric field in the layers depends on the poling direction due to conductivity of the ceramics which was neglected in the modeling of the poling behavior [4]. Here, further investigations are necessary.

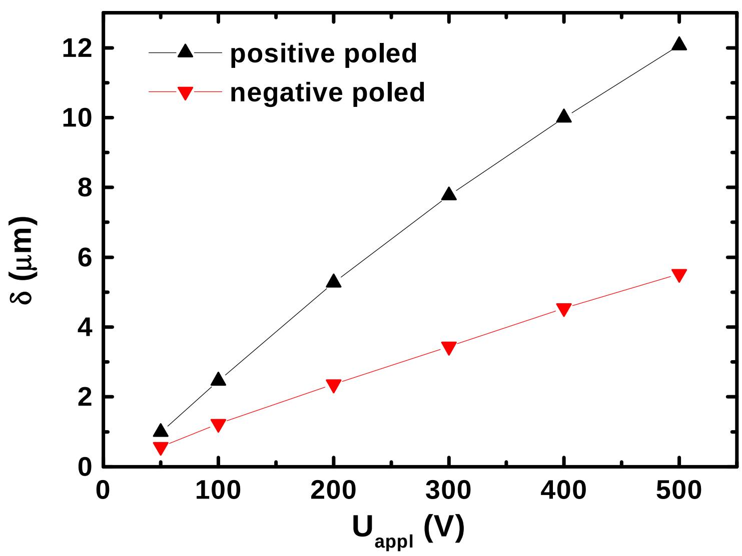

Fig. 7 Bending deflection of monolithic trimorph in dependence of the poling direction

For the trimorph and 4-morph FGM actuators the positive poled specimen show the higher bending deflection (Figures 7and 8). Here the „positive“poling yields a better poling of the BTS7.5 layer. The value of the piezoelectric coefficient d_{31} of this layer increases with the number of layers in the system. The reason seems to be an higher local electric field strength during poling. The higher piezoelectric coefficient also explains the increasing of the maximum deflection with increasing number of layers. For the bimorph we obtained same smaller deflection of the FGM bimorph in comparison with the conventional glued bimorph. We explained this with the gradient of the piezoelectric and dielectric coefficients at the interface. However, we compare FGM samples prepared with different numbers of layers, i.e. with different gradients, the best performance shows the 4-morph with a relatively linear gradient of the tin content. The effect of a „better“poling in a 4-morph is much stronger than the effect of the piezoelectric gradient.
The maximum value of deflection at 500 V was measured of about 0.028 µm/V for a monolithic FGM actuator (4-morph) with a length L = 15 mm and a thickness h = 1,13 mm. That is much higher than the value reported from PNN-PZT based actuators [3].

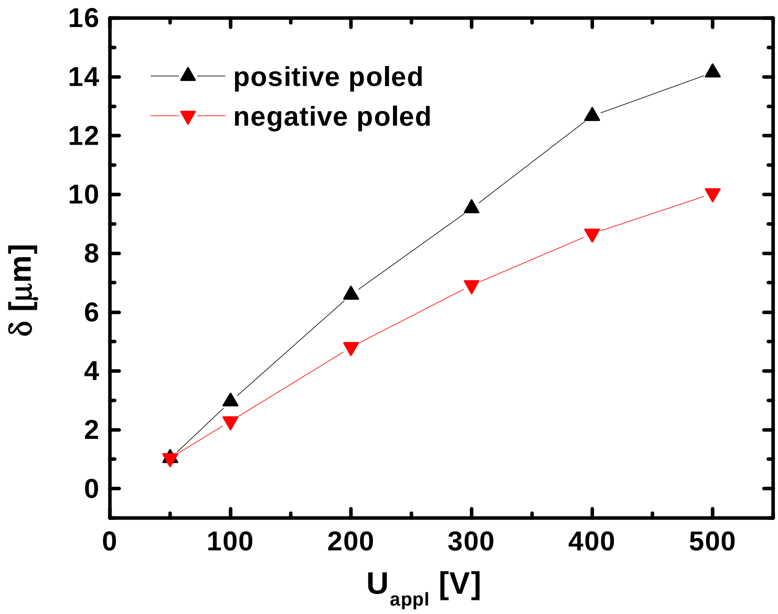

Fig. 8 Bending deflection of monolithic 4-morph in dependence on the applied voltage

Resonance frequencies and damping ratio

For the characterization of the dynamical properties the resonance frequencies and damping ratios of monolithic FGM and the corresponding conventional glued bending actuator were determined.
The frequency of an applied voltage of about 5 V were continuously changed and the amplitude of the deflection was measured by the capacitive sensor. The free length of all samples was 15 mm. A typical resonance curve is shown in Fig. 9. For the conventional glued bimorph consisting of two layers with a defined thickness and constant chemical composition a resonance frequency of about 3150 Hz was measured. From the analytical approximation of the resonance frequency of a one side fixed bending actuator [9]

$$f_s = 0.1615 \frac{h}{L^2} \sqrt{\frac{1}{\rho \, s_{11}^E}} \qquad (3)$$

an average value of the elastic coefficient s_{11} of about $13 \cdot 10^{-12}$ m^2/N was calculated. The influence of the glue was neglected.

In contrast to the quasistatic bending behavior the dynamical properties change in a relatively wide range. The frequency constant

$$N_f = f_s \frac{L^2}{h} \tag{4}$$

of different monolithic FGM actuators was determined between 430 to 600 kHz*mm. A significant dependence on the poling direction or the number of layers could not found. We assume that the form of the gradient (thickness of layers and interface between them) strongly influences the dynamical properties. Probably these parameters are not yet good enough reproduceable.

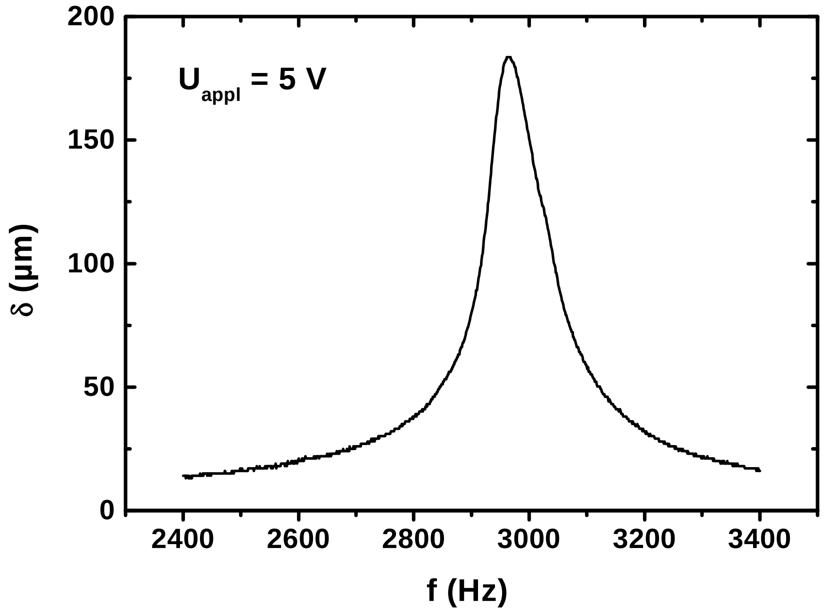

Fig. 9 Resonance curve of a glued bimorph at an applied voltage of 5 V

Additionally, the resonance frequency was measured from the free vibration of the actuator. Therefore, a rectangular signal with a frequency of 40 Hz and a amplitude of 25 V was applied. The deflection response was shown in Figure 10. The results from both methods are in a good agreement.
The damping ratio was determined from the band width of the resonance peak and from the damping of the free vibrations. Here, the results also correspond very well. The damping ratio differs between 0.012-0.028 for bimorph and trimorph FGM actuators and 0.032-0.040 for the monolithic 4-morph ceramics. The values of the damping ratio are comparable with them of glued actuators of about 0.015.

Summary

Functionally Gradient Materials based on Ba(Ti, Sn)O_3 ceramics were prepared. The one dimensional gradient of the tin content along the thickness was varied by using different numbers of powder layers or foils with different Sn content. The distribution of tin after sintering was determined by EPMA. The chemical gradient was transformed to a gradient of the piezoelectric coefficient by a poling process. The poling was more effective than higher the number of layers in the monolithic sample and than „better“ is the functionally gradient. Thus, the monolithic sample prepared from layers shows the best performance. It deflection is much higher than the deflection of a conventional glued bending actuator. Furthermore, a strong dependence of the amplitude of the deflection on the poling direction was found.
The resonance frequencies and damping ratios of monolithic FGM actuators were measured. The dynamical behavior is comparable with glued actuators.

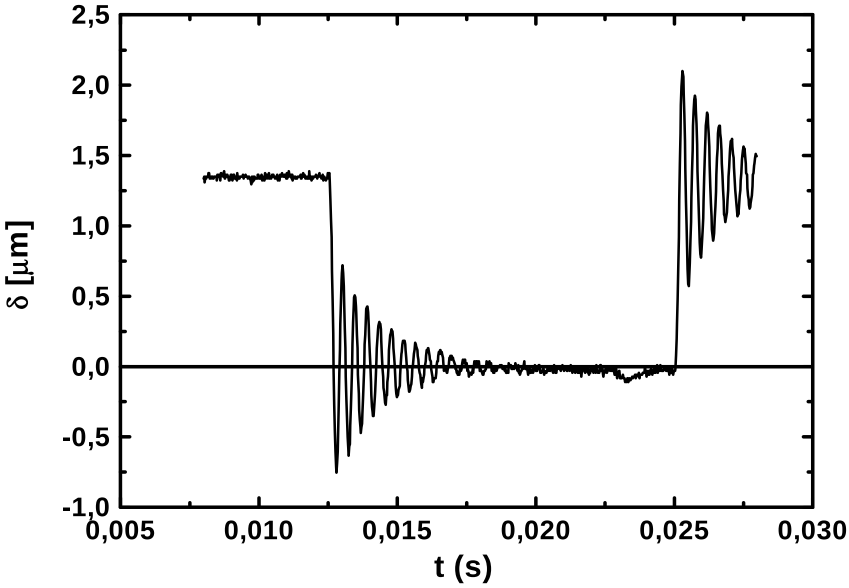

Fig. 10 Free vibration response of a one side fixed monolithic FGM actuator

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